

Perspectives on IONICS and beyond

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Former ARPA-E Program Director (2015 to 2018), led creation and management of the IONICS (Integration and Optimization of Novel Ion-Conducting Solids) program

October 26, 2021

The genesis of IONICS

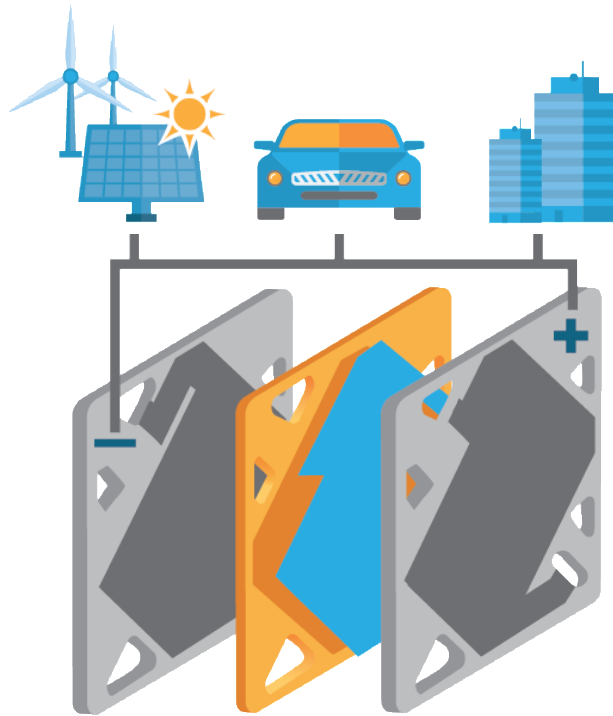
- ▶ IONICS was inspired by gaps in previous device-oriented ARPA-E electrochemical programs.

Solid Ion Conductors for Energy Applications Workshop

Austin Marriott South, Austin, TX
February 19, 2015

Start Time	Speaker	Topic
7:30	REGISTRATION & BREAKFAST	
8:00	Ping Liu, ARPA-E	Welcome, overview of ARPA-E, introductions
	MORNING PRESENTATIONS	
8:30	Paul Albertus, ARPA-E	ARPA-E perspectives on solid ion conductors
9:00	John Goodenough, UT Austin	Solid electrolytes
9:30	Ryoji Kanno, Tokyo Institute of Technology	Solid lithium ion conductor with the LGPS type structure and its application to all solid-state battery
10:00	Break	
10:15	Gerbrand Ceder, MIT	Computational tools to predict the behavior of solid state conduction
10:45	Sossina Haile, Caltech	
11:15	Ramamoorthy Ramesh, UC Berkeley	Interfaces in complex functional oxides
12:00	LUNCH	
	AFTERNOON BREAKOUT SESSIONS	
1:00	Breakout session 1 (topic given at workshop)	
2:30	Read out from session 1 and discussion	
3:00	Break	
3:15	Breakout session 2 (topic given at workshop)	
4:45	Read out from session 2 and discussion	
5:15	Wrap up	
5:30	Adjourn to dinner in Austin	

IONICS: Separators for electrochemical cells



IONICS program mission

Create solid separators for electrochemical cells using solid ion conductors to enable transformational performance and cost improvements in electrochemical cells.

IONICS: a transformational component demonstrated in the device context

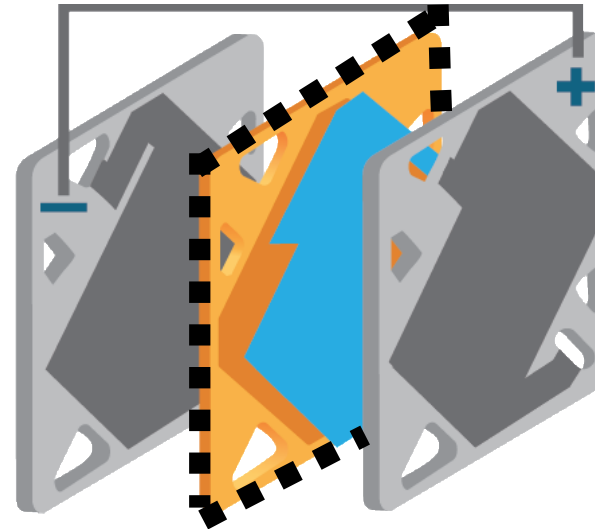
Typical ARPA-E program



Device metrics:

W/kg, Wh/kg, \$/kW, \$/kWh, durability, mA/cm² at a given V, etc.

IONICS program

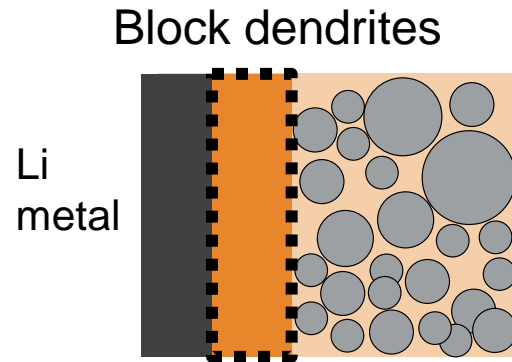


Component metrics:

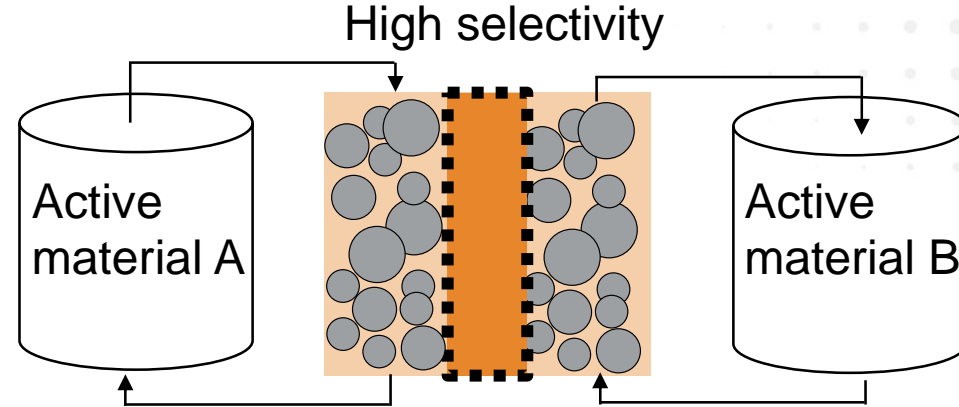
Selectivity, stability, separator and interfacial ASR, dendrite resistance, \$/m².

IONICS focuses on three types of separators for three cell platforms

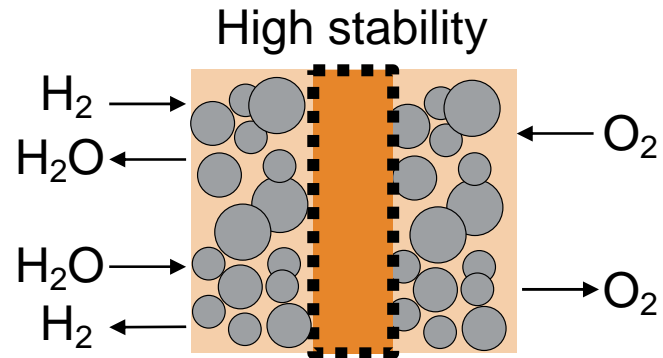
Category 1: Li metal battery



Category 2: Flow battery



Category 3: H₂/O₂ alkaline devices



Why are we still talking about rechargeable lithium metal batteries, seven (in the case of IONICS) and ~fifty (for the field in general) years later?



Contents lists available at ScienceDirect

Journal of Neuroscience Methods

journal homepage: www.elsevier.com/locate/jneumeth



Basic Neuroscience
Review

DARPA-funded efforts in the development of novel brain–computer interface technologies



Robbin A. Miranda^a, William D. Casebeer^b, Amy M. Hein^c, Jack W. Judy^d, Eric P. Krotkov^e, Tracy L. Laabs^c, Justin E. Manzo^f, Kent G. Pankratz^f, Gill A. Pratt^g, Justin C. Sanchez^b, Douglas J. Weber^b, Tracey L. Wheeler^h, Geoffrey S.F. Ling^{b,*}

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^h System Planning Corporation, 3601 Wilson Boulevard, Arlington, VA 22201, USA

H I G H L I G H T S

- DARPA's programs foster multi-disciplinary collaborations.
- DARPA's BCI programs span four major challenges: detect, emulate, restore, & improve.
- Aims: restore function after injury; improve performance of healthy individuals.

A R T I C L E I N F O

Article history:

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A B S T R A C T

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“DARPA has funded innovative scientific research and technology developments in the field of brain-computer interfaces (BCI) *since the 1970s.*”

Takeaways from these recollections

- ▶ Battery R&D requires time and benefits from investments by multiple programs and agencies, and the private sector.
- ▶ For an ARPA-E size and duration program, focusing on a single component (in a device context) worked for IONICS.

Status and challenges in enabling the lithium metal electrode for high-energy and low-cost rechargeable batteries

Paul Albertus^{1*}, Susan Babinec¹, Scott Litzelman² and Aron Newman²

Enabling the reversible lithium metal electrode is essential for surpassing the energy content of today's lithium-ion cells. Although lithium metal cells for niche applications have been developed already, efforts are underway to create rechargeable lithium metal batteries that can significantly advance vehicle electrification and grid energy storage. In this Perspective, we focus on three tasks to guide and further advance the reversible lithium metal electrode. First, we summarize the state of research and commercial efforts in terms of four key performance parameters, and identify additional performance parameters of interest. We then advocate for the use of limited lithium ($\leq 30 \mu\text{m}$) to ensure early identification of technical challenges associated with stable and dendrite-free cycling and a more rapid transition to commercially relevant designs. Finally, we provide a cost target and outline material costs and manufacturing methods that could allow lithium metal cells to reach $100 \text{ US\$ kWh}^{-1}$.

Lithium-ion batteries are transforming the transportation and grid sectors. Their scale up is truly historic: Li-ion is now the only rechargeable battery other than lead acid produced at $>5 \text{ GWh yr}^{-1}$, with a worldwide manufacturing expansion reaching hundreds of GWh yr^{-1} over the next five years^{1,2}. Li-ion battery packs achieve long cycle life (in the thousands), high charge/discharge rates ($>1 \text{ C}$), high energy content (specific energy of $\sim 150 \text{ Wh kg}^{-1}$ and energy density of 250 Wh L^{-1}), and low capital costs ($<300 \text{ US\$ kWh}^{-1}$)^{3,4}. However, the present Li-ion material platform (a graphite negative electrode coupled with a metal oxide positive electrode) is not expected to reach the US Department of Energy's (DOE) electric vehicle pack goals of 235 Wh kg^{-1} , 500 Wh L^{-1} and $125 \text{ US\$ kWh}^{-1}$ (ref. 5). The intercalation mechanism that fundamentally enables the excellent cycling of Li-ion also places an upper limit on energy content because of the weight and volume of the hosts into which Li^+ intercalates. Thus, there remains an acute need for higher-energy alternatives to the Li-ion material platform^{6–8}. Replacing the graphite electrode with lithium metal (Fig. 1), which results in a $\sim 35\%$ increase in specific energy and $\sim 50\%$ increase in energy density at the cell level, provides a path to reach those goals, especially if the introduction of lithium metal is combined with reduction of the liquid electrolytes, which impose both safety and thermal management mass and volume requirements at the pack level^{9–12}.

Stable cycling of lithium metal requires high Coulombic efficiency, a low and stable resistance, and the avoidance of lithium dendrites. To meet these requirements a myriad of approaches have been pursued (starting in the 1960s), most of them focused on the use of solid, liquid, composite and other electrolytes^{13–19}. Vapour-deposited lithium phosphorus oxynitride (LiPON) solid separators, and thin-film cells based thereon, were a major advance in the late 1990s and remain the definitive example of stable cycling of lithium metal at room temperature^{20,21}. Another key advance came with the development of polyethylene oxide polymer electrolytes, which unfortunately require elevated temperature ($\sim 80^\circ\text{C}$) and

cathodes operating at $<4 \text{ V}$ versus Li (refs 22,23). Because neither of these material platforms currently surpass Li-ion for vehicle or grid applications, development efforts continue. Unfortunately, such efforts are hampered by the lack of a systematic understanding — incorporating both theory and experimental validation — of the conditions under which lithium dendrites do and do not form^{18,24–29}. As just one example, the physical form of lithium dendrites (or more generically, lithium penetrations) differs significantly based on the medium through which they are growing: lithium penetration through a polycrystalline inorganic solid electrolyte has a much different morphology than growth through a liquid^{30,31}. Regardless of the approach, challenges for high-rate, high-capacity cycling of a metal electrode are profound. Metal dendrites are known to grow in numerous systems, and observations of Ag dendrite growth through a solid inorganic ion conductors appear to go back nearly 100 years^{32,33}. In the case of lithium, dendrites are known to grow through many classes of matter, including liquids, polymers, glasses, and polycrystalline inorganic solid electrolytes^{19,28,30,34}.

In this Perspective, we provide a new figure that includes four parameters of critical importance for the practical development of a lithium metal electrode, and use this figure to summarize the status of both academic and commercial efforts. We find that although impressive results have been achieved for subsets of the four critical parameters, none have met targets for all four, and we suggest researchers focus their attention on the complete set of critical parameters when cycling cells. Perhaps the biggest gap in the parameters used for lithium metal cycling today is a large excess of lithium — far more than could be included in a high-energy cell — and an amount that makes it difficult to perform the careful experiments needed to ensure stable, dendrite-free cycling. In particular, we believe that numerous reports of stable lithium metal cycling in symmetric Li/Li cells are in fact the result of 'soft shorts' (stable electronic connections between the electrodes). These carry electronic current; fortunately, the use of a limited amount of lithium ($\leq 30 \mu\text{m}$, now available commercially) can help identify soft shorts

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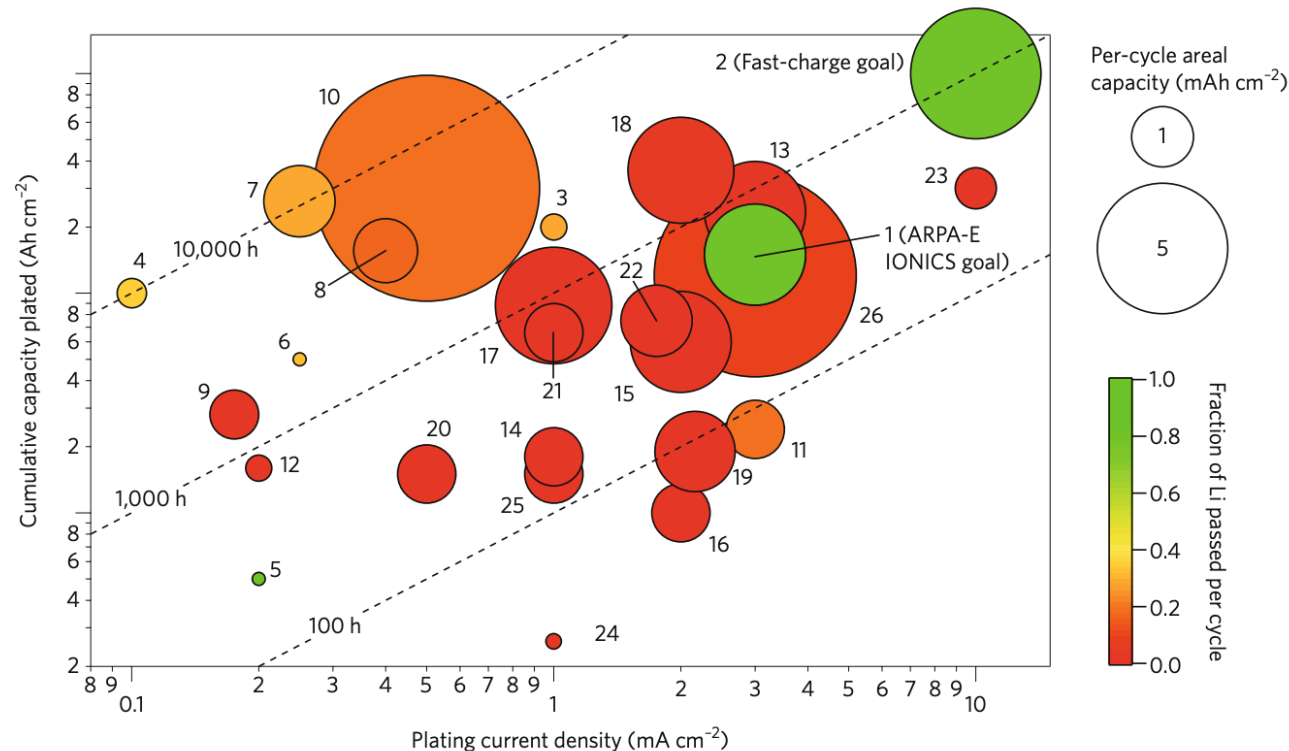
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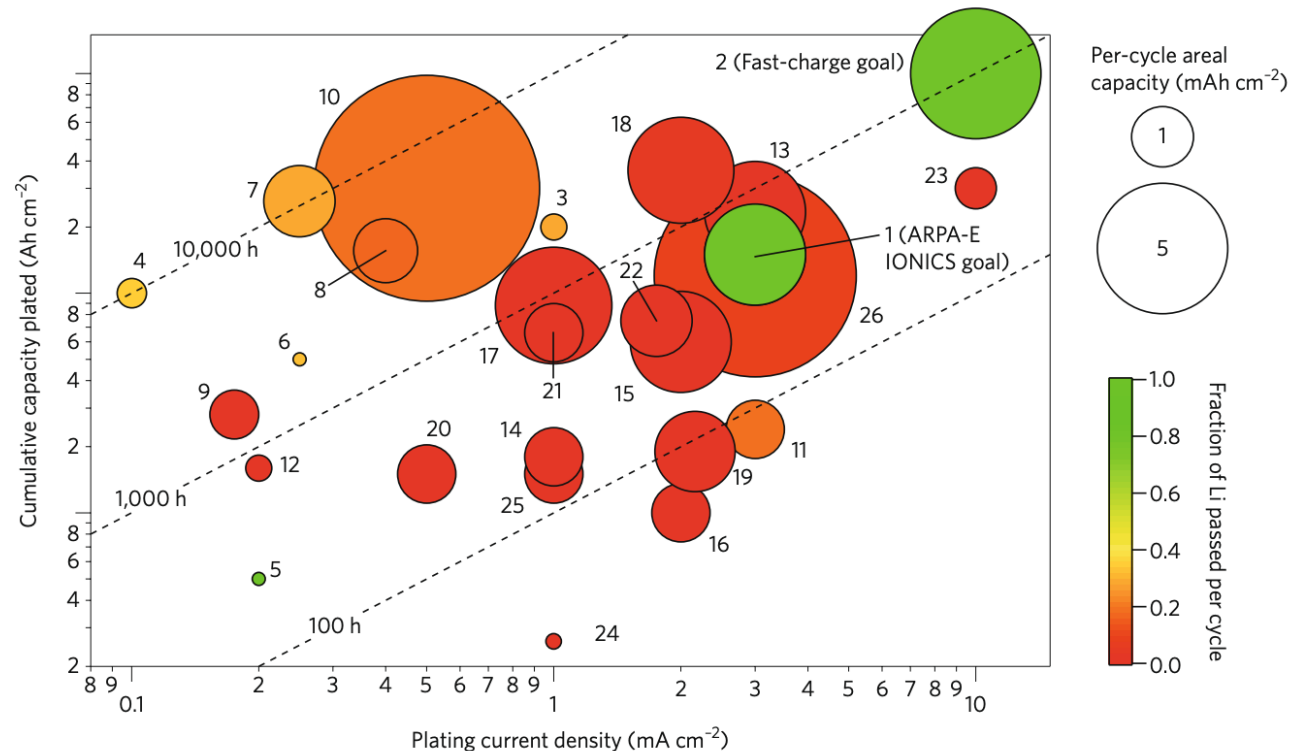
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350 kW
Electrify
America
charge

A possible future target:
350 kW chargers
75 kWh battery pack
4.67 C-rate
For 5 mAh/cm^2 , $>20 \text{ mA/cm}^2$

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Fast charge Li metal at $>20 \text{ mA/cm}^2$??

Crazy good or crazy bad?

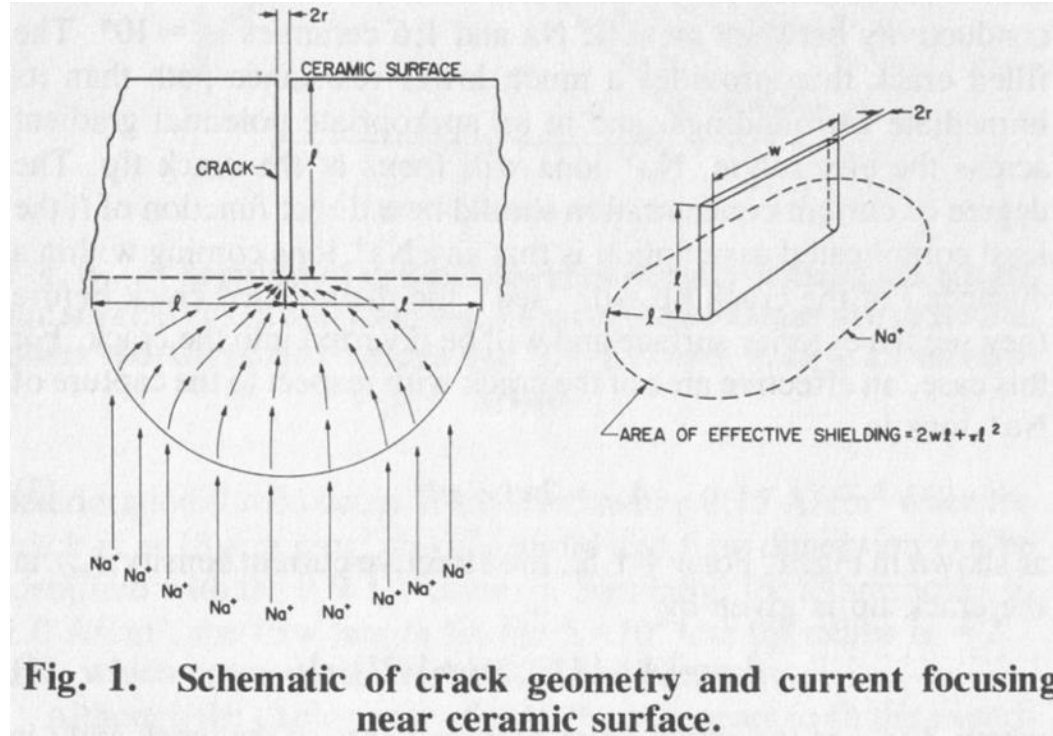
Molten Na, Li with solid electrolytes cycle at >100 s of mA/cm²

A Model for Degradation of Ceramic Electrolytes in Na-S Batteries

R. H. RICHMAN and G. J. TENNENHOUSE

Scientific Research Staff, Ford Motor Company, Dearborn, Michigan 48121

1975



Molten Na, Li with solid electrolytes cycle at $>100\text{s}$ of mA/cm^2

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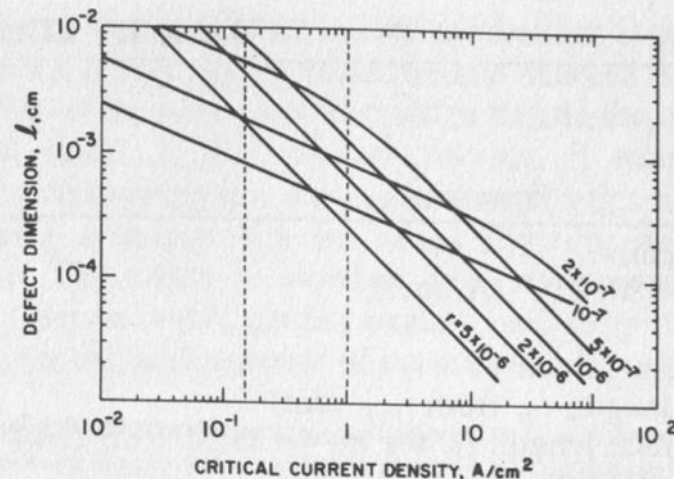


Fig. 4. Variation of critical current density with crack length for several crack thicknesses. *Vertical dashed lines* are drawn at observed threshold values for 9.9-1.1 and 9.25-0.25 electrolytes.

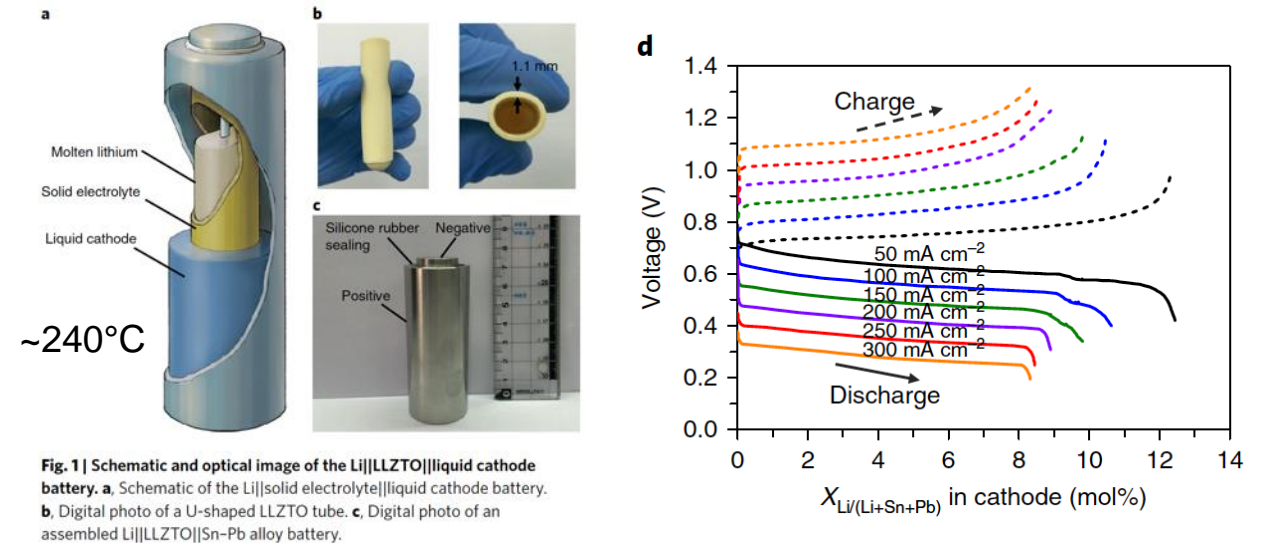
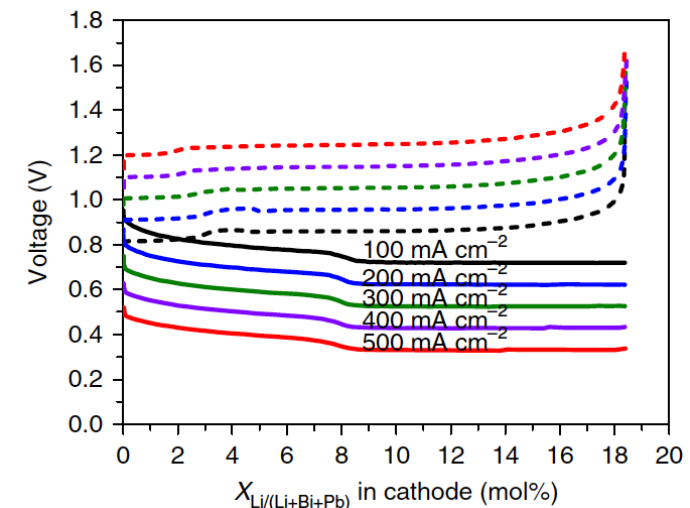
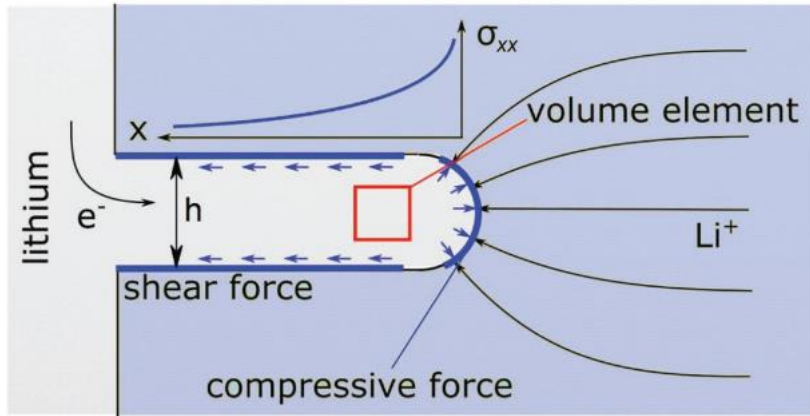


Fig. 1 | Schematic and optical image of the Li||LLZTO||liquid cathode battery. **a**, Schematic of the Li||solid electrolyte||liquid cathode battery. **b**, Digital photo of a U-shaped LLZTO tube. **c**, Digital photo of an assembled Li||LLZTO||Sn-Pb alloy battery.

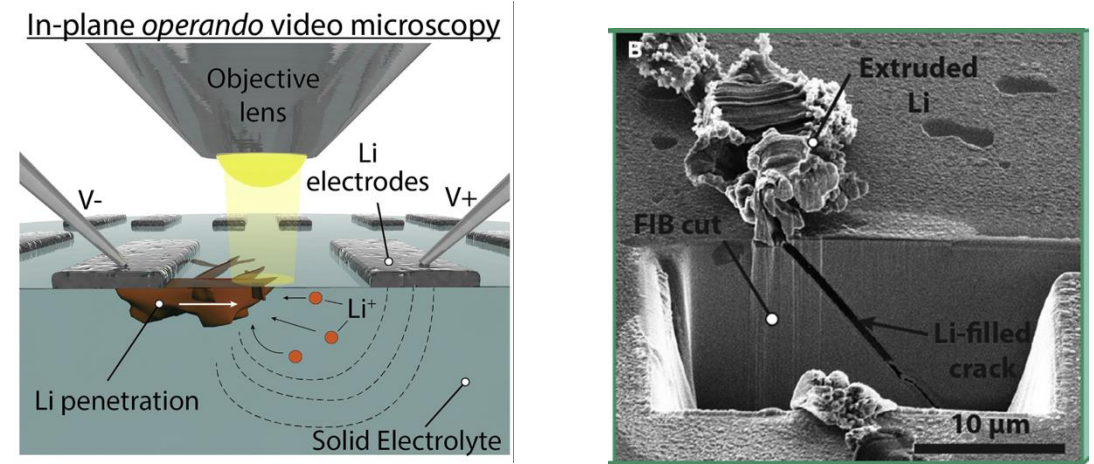


The last 5 years have produced outstanding scientific work in the area of solid electrolytes and lithium metal

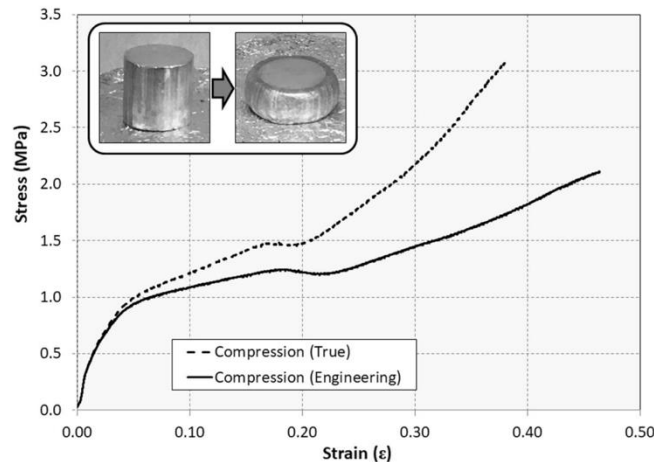
Electrodeposition driven fracture mechanism (2017)



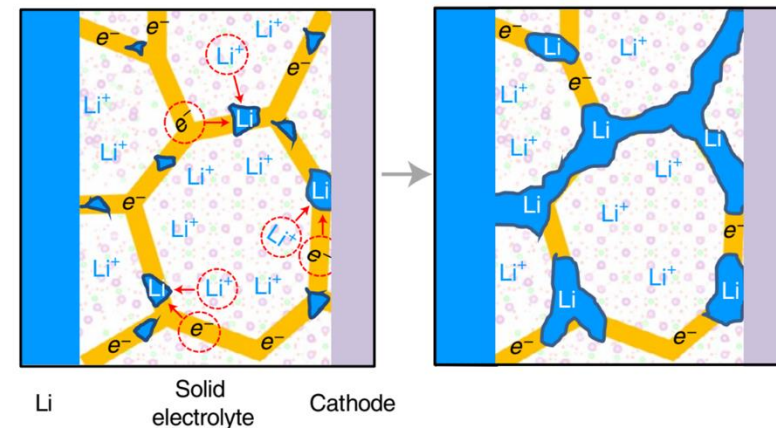
Microscopy of Li penetration (2020)



Mechanical behavior of Li metal (2019)



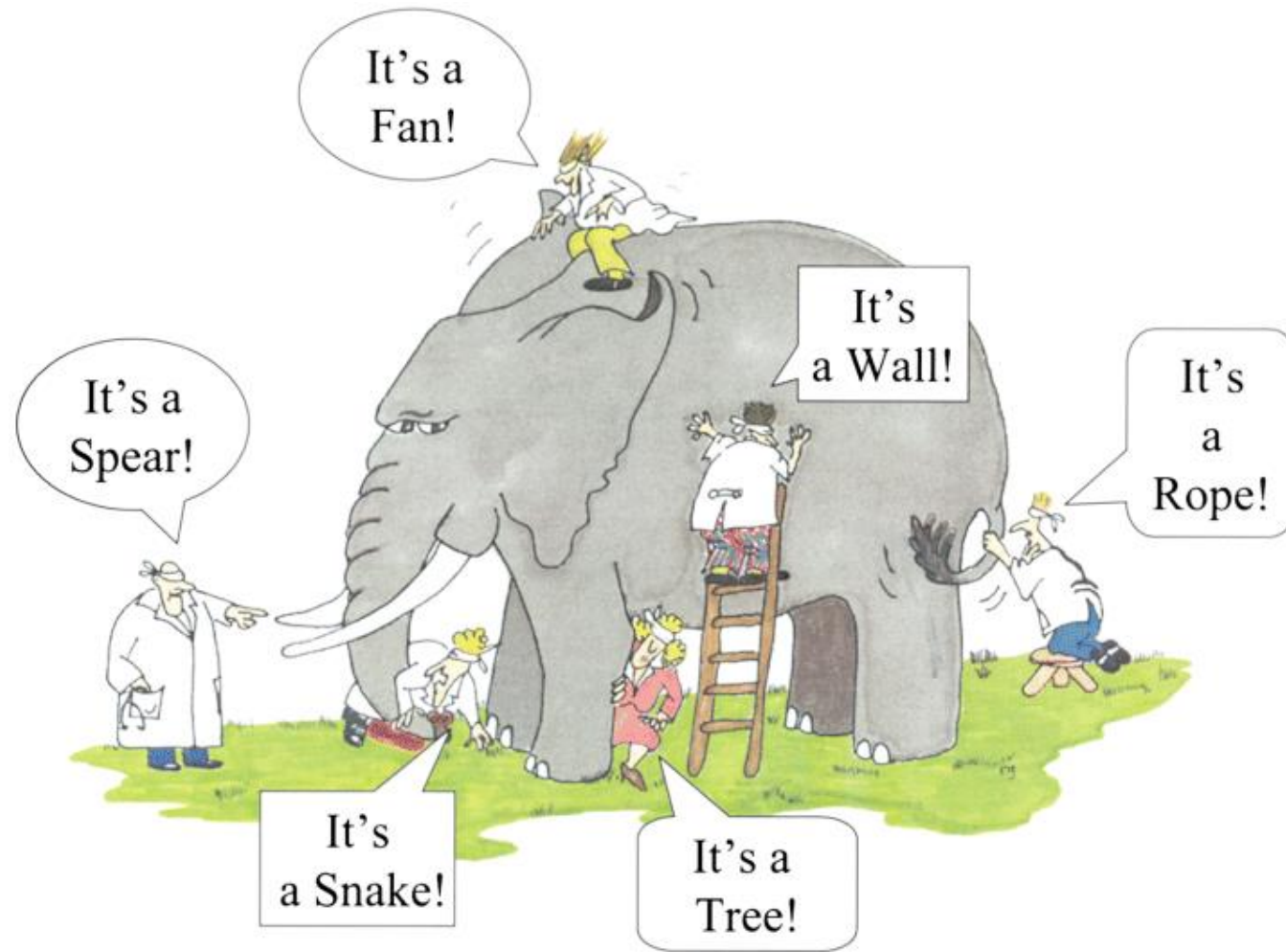
New mechanisms (e.g., electronic conductivity) (2019)



Porz et al, Advanced Energy Materials, 2017.
Masias et al, J Mater Sci, 2018.

Kazyak et al, Matter, 2020.
Liu et al, Nat Mater, 2021.

There is still limited and fragmented knowledge in quantifying performance limits of lithium metal with a solid-state separator



There are lots of creative ideas informed by the improving science

- ▶ 3D anodes to reduce the interfacial current density.
- ▶ Making a “perfect” interface (in geometrical and other properties).
- ▶ Warming the battery during fast charge (already done for Li-ion today).
- ▶ Interlayers.
- ▶ Li metal alloys.
- ▶ Building cells without metallic Li.
- ▶ Processing.
- ▶ ...

Thank you!
